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Ammonia		
Sulfur Oxides		

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REVIEW OF U.S. NO_x ABATEMENT TECHNOLOGY

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ABSTRACT

Research and development of nitrogen oxide (NO_x) control techniques in the United States has included a variety of combustion related techniques as well as selective catalytic reduction (SCR) of NO_x with ammonia (NH_3). Current national emission standards for both utility and industrial boilers will require either low NO_x burners or combustion modifications to achieve compliance. The State of California has more stringent regulations that, in some cases, require the use of SCR to achieve compliance; however, there have been no commercial applications to utility boilers. Results from pilot unit testing of SCR have allowed the costs of full scale systems to be estimated. Current research is directed toward processes that remove NO_x and sulfur oxides (SO_x) simultaneously.

INTRODUCTION

The United States has recognized for several years the need to control nitrogen oxide (NO_x) emissions and has implemented NO_x control programs for both stationary and mobile sources. This paper will describe research efforts, demonstration projects, and commercial applications designed to control NO_x from stationary sources.

Research efforts directed toward NO_x control have included both modifications to the combustion process and post-combustion NO_x removal as well as simultaneous control of NO_x and SO_x. Most of the research has involved modifying the combustion process since this approach is much less expensive than post-combustion techniques. Initial research in the early 1970's addressed combustion modifications such as overfire air, staged combustion, and flue gas recirculation. This led to the

development of low NO_x burners which have been applied to many boilers. Other approaches to the combustion control of NO_x have been the development of fluidized bed combustion (FBC), limestone injection multistage burners (LIMB), in-furnace NO_x reduction (reburning), and precombustors. Post-combustion techniques have also been tested and applied: the most popular are selective catalytic reduction (SCR) and selective noncatalytic reduction of NO_x with ammonia (NH_3).

As a result of this research and development effort there have been many commercial applications of NO_x control technology, and many others are underway. Of these NO_x control techniques, the most widely applied are combustion modification techniques and low NO_x burners since these are the most cost effective and can meet the requirements of federal NO_x emission regulations. SCR was tested at several coal-fired boiler locations and the results used to perform economic analyses for full scale applications. At the present time, there has been only one full scale application of SCR: a demonstration system that will cease operation next year. Current research and development activities are focused primarily on simultaneous NO_x/SO_x control processes.

NO_x ISSUES IN THE UNITED STATES

The Environmental Protection Agency (EPA) has limited the amount of NO_x that can be emitted by new utility and industrial boilers larger than 73.3 MJ/sec heat input. Emission regulations have also been proposed for new industrial boilers larger than 29.3 MJ/sec heat input. The emission limits for new utility and industrial boilers in the U.S. are shown in Table 1 /1,2,3/. All of these emission levels can be achieved through application of commercially available combustion technology.

The State of California is an exception in that new utility and industrial boilers are required to meet emission levels that can only be attained through the use of post-combustion treatment. As a result, there are several post-combustion systems applied in California.

CURRENT STATUS OF NO_x CONTROL DEVELOPMENT

Combustion Modifications and Low NO_x Burners

Several types of low NO_x burners have been developed and many have been applied to utility boilers. The 1979 emission standards for utility boilers require low NO_x burners on all new boilers; however, there have not been many new boilers built so applications are limited. The most widely applied is the Babcock & Wilcox dual-register burner which has been applied to 34 boilers with a combined generating capacity of 20,000 MW /4/. These are first generation design burners and, in general, burners in this classification have demonstrated NO_x control levels of up to 60 percent. Since these initial designs, advanced low NO_x burners have been developed, and some have been installed in utility boilers. Table 2 summarizes EPA sponsored tests of several burner designs on a variety of boiler designs. These results indicate that first generation designs achieve NO_x emissions of 163 to 245 ng/J. The advanced designs can achieve about the same level of NO_x control and will outperform the early designs when the low NO_x burners are included as part of the boiler design. The last three tests shown in Table 2 are long term tests in which the effect of the boiler load and other variables can be seen. The range of daily averages can vary greatly from the 30 day average and, therefore, it is difficult to predict performance from single point tests. While the long term emissions from boiler C are high when compared to the others, it should be noted that this is an old unit designed with a small firebox which is not used in boilers designed for low NO_x.

Tests have been performed by Southern California Gas Company on two full-scale engines to evaluate the effectiveness of SCR with ammonia injection and nonselective catalytic reduction (NSCR) on NO_x emissions from natural-gas-fired stationary internal combustion engines. The NSCR system reduced NO_x from 1700 ppm, at 15 percent O_2, to 559 ppm, and total organic levels were also reduced significantly. However, both ammonia and cyanide emissions were increased across the catalyst. Results on the SCR tests were better. NO_x was reduced from 1470 ppm to

310 ppm at 15 percent O_2. Also, EPA is running a 4000 hour test of the SCR system on a diesel engine. After 1000 hours of operation, a NO_x reduction exceeding 90 percent was achieved.

Advanced Combustion Modification Technology

Several companies and agencies have recently supported research to investigate the potential for obtaining NO_x reduction via in-furnace NO_x reduction, or reburning. This technology utilizes 15 to 40 percent of the fuel to form a second combustion zone which is usually operated fuel-rich. NO_x formed in the primary combustion zone is reduced to N_2 and H_2O when it passes through the reducing zone. Key parameters are residence time, temperature, and fuel nitrogen in the reburn fuel. Tests performed by EPA and their contractors have shown that NO_x can be reduced by up to 85 percent under optimum conditions. More typically, Japanese tests on full-scale combustors have shown that 50 percent NO_x reduction is routinely achievable.

Another advanced combustion modification technology for NO_x control is the precombustor. This is an advanced burner design which results in a fuel-rich primary zone at very high temperature. The excess air for burnout is ended downstream at lower temperatures. EPA has sponsored the development of a heavy-oil low-NO_x burner that is capable of operating with NO_x emissions of 75 ppm. This technology is also being evaluated for coal-fired burners, also known as slagging burners. Avco, Coal Tech, Rockwell, and TRW are all developing burners which utilize this technology. EPA is evaluating the potential for simultaneous SO_x control via sorbent injection.

Flue Gas Treatment

Post combustion NO_x control, especially selective catalytic reduction (SCR), has been the subject of three pilot unit demonstrations, one full scale demonstration, and several detailed engineering economic assessments. The EPA has sponsored two pilot unit demonstrations at the 0.5 MW scale using flue gas extracted from coal-fired boilers. Two different processes were tested. One, the Hitachi Zosen process, is a NO_x-only process that injects NH_3 into the

flue gas and passes it through a "honeycomb" type of monolithic catalyst. The pilot unit was located at Georgia Power's Mitchell Station. The second process tested was the Shell Flue Gas Treatment process which removes both SO_x and NO_x from the flue gas using a copper oxide acceptor material. In this process, the acceptor is regenerated using hydrogen, and an SO_2-rich byproduct stream is produced that can be refined to pure SO_2, H_2SO_4, or elemental sulfur. This process was tested at Tampa Electric's Big Bend Station.

The results of these pilot unit tests are summarized in Table 3. The Hitachi Zosen process was able to achieve a long term NO_x control level of 90 percent. The Shell process achieved 90 percent SO_x removal, but only 70 percent NO_x removal. Additional catalyst would probably allow the Shell process to achieve 90 percent removal of both SO_x and NO_x.

The results of these tests were used to estimate the costs of applying either process to a 500 MW, coal-fired boiler. For the Hitachi Zosen process the capital and annual costs were estimated to be $44/kW and 2.6 mills/kWh, respectively. The costs of the Shell process were estimated to be $166/kW and 8.4 mills/kWh /12/. Other economic analyses have been performed comparing the Shell process with the combination of conventional SCR and a limestone-based, flue gas desulfurization (FGD) system. The results indicated that the combination of SCR and limestone FGD is more economical.

A third pilot unit demonstration of SCR was sponsored by the Electric Power Research Institute (EPRI) at Public Service of Colorado's Arapahoe Station. This was a 2.5 MW scale system that used the Kawasaki Heavy Industries (KHI) SCR technology. The demonstration is summarized in Table 4. The inlet NO_x concentration was very high, 950 ppm, yet the system was able to achieve 80 to 90 percent NO_x removal; however, this resulted in high NH_3 concentrations in the treated flue gas /13/. The results of the pilot unit tests were used to perform detailed costing studies, and these results will be presented later in this paper.

A full scale, 107.5 MW, demonstration of the KHI SCR system is being conducted by Southern California Edison at their Huntington Beach Station. A description of this demonstration is contained in Table 5.

The boiler fires oil 30 percent of the time and gas 70 percent of the time. With both fuels, the SCR system is capable of 90 percent NO_x removal over a wide load range. Initially there were problems with the process instrumentation with either SO_3 or $(NH_4)_2SO_4$ condensing; however, these problems have been overcome. This is a retrofit system and is relatively expensive at $79/kW. The SCR system will cease operation during March 1985 /14/.

The results of these demonstrations have been used to perform detailed economic analyses of SCR systems. Two similar studies, one by EPA and the other by EPRI, were completed in late 1984. The EPA study was prepared by the Tennessee Valley Authority and considered three separate cases. These are presented in Table 6. The objective was to determine the overall costs of pollution control on a 500 MW boiler. Different combinations of SO_2 and particulate control were used with SCR to determine a range of capital and annual costs. The results of this study are shown in Table 7 in terms of the absolute cost of SCR and as a percentage of the whole pollution control system. Capital costs ranged from $83.7 to $100.2/kW and levelized annual costs ranged from 13.0 to 15.8 mills/kWh /15/.

A similar cost study was conducted by the Electric Power Research Institute using the results of their pilot unit experience. Two coals, three inlet NO_x concentrations, and three NO_x removal levels were considered in order to get a range of costs. The results are presented in Table 8. The study concludes that the capital cost of SCR is approximately $54 to $98/kW depending on the application. Similarly the annual costs are 5.7 to 14.9 mills/kWh. The costs for the low sulfur coal case are considered to be more accurate than those for high sulfur coal since the pilot unit results were obtained with low sulfur coal /16/.

The results of the two economic analyses compare favorably, although there is a wide range between the minimum and maximum costs. This range is the result of application specific factors and is to be expected. These two studies are compared for the use of 80 percent NO_x removal from a 500 MW boiler firing low sulfur coal in Table 9. It can be assumed that the capital costs of retrofit systems will be from one to three times those of new systems.

These tests and evaluations represent the current extent of research and development of SCR for utility boiler applications in the U.S. There are no applications at the present time since the Federal regulations can be met through application of combustion modifications and/or low NO_x burners. SCR is a useful NO_x control technique in situations where maximum NO_x control is necessary. This is the case in the State of California where the NO_x emission limitations imposed by the local regulatory agencies are more stringent than the Federal standards.

There have been several commercial applications of SCR in California to industrial boilers, refinery heaters, and stationary engines. A list of these applications is shown in Table 10 /17/. SCR has been considered for application to oil field steam generators, but has not yet been applied. California is also requiring SCR on gas turbines used at cogeneration facilities. There have been no applications at the present time; however, three permit requests have been denied because the proposed facility did not include SCR. One suggested emission level for turbines in California is 25 ppm measured at 15 percent O_2.

CURRENT STATUS OF SIMULTANEOUS NO_x/SO_x CONTROL DEVELOPMENT

Fluidized Bed Combustion

Another combustion approach to NO_x control is fluidized bed combustion (FBC). This technique is the result of long term research and development efforts by the Environmental Protection Agency (EPA), the Electric Power Research Institute (EPRI), the Tennessee Valley Authority (TVA), the Department of Energy (DOE), and others. Several pilot units have been built and tested with promising results. FBC is now emerging as a commercial technology, and there are currently three utilities planning to install full scale FBC boilers. These are listed in Table 11. The Northern States Power unit is interesting in that an existing boiler will be modified for fluidized bed operation. Based on pilot unit testing at the 20 MW scale, FBC is expected to reduce NO_x emissions by 20-30 percent. In addition, FBC can control SO_2 emissions by as much as 90 percent.

Limestone Injection Multistage Burners

Currently, the most extensive area of combustion related research into NO_x control systems is the development of limestone injection multistage burners (LIMB). These have been developed over the last 5 years, and pilot unit testing is essentially complete. The low NO_x burner used is the distributed mixing burner (DMB) developed by EPA. A full scale demonstration has been initiated with costs to be shared by EPA and the utility, Ohio Edison. The demonstration will be on a 105 MW boiler, the Edgewater Station, firing high sulfur coal, and will last 4.5 years with 1 year of actual testing. The primary objectives are to demonstrate 50 to 60 percent control of both NO_x and SO_x, and determine the impacts of LIMB on the operation of the boiler. The retrofit burner design is currently at the engineering stage, and operation will begin in about 2 years. Also, a full scale demonstration of LIMB on a tangentially fired boiler is being considered.

Flue Gas Treatment

There are several research projects in the U.S. examining post-combustion NO_x control. The processes being considered are those that remove both NO_x and SO_x, and are not commercially established. The Department of Energy is sponsoring several bench scale and pilot unit projects. The processes that are being tested at the pilot unit level are listed in Table 12. These processes or variations of them should be familiar to those who have followed the development of post-combustion NO_x control.

The fluidized copper oxide bed process is essentially a fluidized bed concept of the Shell process. Current work is complete, and a larger scale pilot unit is being considered. The electron beam concept is being examined by itself as a SO_x/NO_x process and in combination with a spray dryer. Testing of the combination system will be completed soon. The 6 MW electron beam pilot unit is scheduled to begin operation in 1985. The SULF-X process was originally designed as an SO_x removal process; however, the pilot unit has been modified for SO_x/NO_x operation and will begin operation soon /18/.

SUMMARY AND CONCLUSIONS

The preceding discussion has covered the current state of research and development of NO_x controls for stationary sources in the U.S. It has also covered the degree to which each technology has been applied. From this, the following conclusions can be drawn:

- Combustion modifications and low NO_x burners have been shown to be capable of limiting NO_x emissions from 163 to 245 ng/J.
- Advanced versions of these techniques can achieve these levels and lower when applied to new boilers.
- These NO_x control techniques are sufficient to meet the current standard for utility boilers and the proposed standard for industrial boilers.
- Research on advanced combustion concepts for NO_x control using LIMB for simultaneous NO_x/SO_x control, is on-going and will continue for several years.
- An assessment of the cost and feasibility of application of SCR to coal-fired and utility boilers has been completed.
- The pilot unit demonstrations of SCR indicate that 90 percent NO_x control was technically achievable on coal-, oil-, or gas-fired boilers.
- For a 500 MW boiler firing low sulfur coal, the costs of operating an SCR system at 80 percent NO_x control are \$79.1-\$100.2/kW (capital) and 11.7-15.8 mills/kWh (annual).
- SCR has been applied in the U.S. to industrial boilers and refinery heaters and stationary engines in California.
- Current post-combustion NO_x control research in the U.S. is focused on uncommercialized processes that have both NO_x and SO_x removal capability.

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TABLE 1. SUMMARY OF CURRENT NO_x EMISSION REGULATIONS FOR BOILERS IN THE UNITED STATES /1,2,3/

	NO_x emission limits, ng/J		
	Utility and industrial boilers[1]	Utility boilers[2]	Industrial boilers (proposed)[3]
Gaseous fuels			
Coal derived		210	
All other	86	86	43
Liquid fuels			
Coal derived		210	
All other	130	130	
Distillate oil			43
Residual oil			
Fuel nitrogen ≤0.35 wt. %			129
Fuel nitrogen ≥0.35 wt. %			172
Solid fuels			
Subbituminous coal	300	210	
Bituminous coal	300	260	
Anthracite coal		260	
Mass feed stoker			215
Spreader stoker			258
Pulverized coal			301
Lignite	260-340	260	258

[1] >73.3 MJ/sec heat input, effective 1971.

[2] >73.3 MJ/sec heat input, effective 1979.

[3] >29.3 MJ/sec heat input, not effective yet.

TABLE 2. RESULTS OF FULL SCALE LOW NO_x BURNER DEMONSTRATIONS /5,6,7,8,9/

	Single point tests		Long term tests		
Boiler	A	B	C	D	E
MW output	60	360	20	400	265
Coal	Bitum.	Subbitum.	Bitum.	Bitum.	Bitum.
Type of firing	Tangential	Front wall	Front wall	Tangential	Front wall
Date operational	Before 1971	1976	1965	1981	1979
Stack O_2, %	4.5	9.7	4.0	4.2	5.5
Low NO_x burner	Tangential	Controlled flow, split flame	Distributed mixing	Tangential, offset air	Dual register
Overfire air	Yes	No	Yes	Yes	No
Conventional or advanced design	Conventional	Advanced	Advanced	Advanced	Conventional
New or retrofit	Retrofit	Retrofit	Retrofit	Retrofit	New
Days of continuous data	Single point	1	30	120	69
Average NO_x emissions, ng/J	245	176	211	176	163
Range of daily averages, ng/J	-	-	185-288	82-241	129-206

A = Richmond Power and Light Company, Whitewater Station
B = Public Service of New Mexico, San Juan Station
C = Western Illinois Power Company, Pearl Station
D = Utah Power & Light, Hunter Station
E = Southern Indiana Gas and Electric Company, A.B. Brown Station

TABLE 3. RESULTS OF EPA PILOT UNIT TESTS AND ECONOMIC PROJECTIONS /12/

	Hitachi Zosen	Shell Flue Gas Treatment
Pilot unit		
Size, MW	0.51	0.5
Flue gas flow rate, Nm^3/hr	1600	1600
NO_x removal, %	90	70[1]
SO_x removal, %	-	90
Estimates for a 500 MW coal-fired boiler[2]		
Energy usage, % of boiler capacity	3.8	5.0
Costs (basis: 1980)		
Capital, $/kW	44	168
Annual, mills/kWh	2.6	8.4

[1] 90 percent probably achievable with additional acceptor material.

[2] Basis: 90 percent NO_x removal, 90 percent SO_2 removal.

TABLE 4. DESCRIPTION OF SCR PILOT UNIT OPERATED BY THE ELECTRIC POWER RESEARCH INSTITUTE /13/

Size, MW	2.5
System vendor	Kawasaki Heavy Industries
Inlet NO_x, ppm	950
Inlet SO_x, ppm	450
NO_x removal, %	80 to 90
Residual NH_3, % of inlet	2.5 to 4.5
Coal	Low sulfur western

TABLE 5. FULL SCALE SCR DEMONSTRATION AT SOUTHERN CALIFORNIA EDISON'S HUNTINGTON BEACH STATION /14/

Boiler size, MW	107.5
Fuel	Oil - 30% of time Gas - 70% of time
Flue gas flow rate, m^3 at 399°C	850,000
NO_x concentration, ppm	180 - 280 (oil firing)
SCR system vendor	Kawasaki Heavy Industries
NO_x removal, %	90
Residual NH_3, ppm	<10
Load variation, MW	20 to 107.5
Start-up	March 1982
Shutdown (scheduled)	March 1985
Installed cost (retrofit)	$8,500,000 ($79/kW)

TABLE 6. BASIS FOR ECONOMIC ANALYSES OF THE TVA/EPA STUDY /15/

	Case 1	Case 2	Case 3
Coal and boiler conditions			
Coal	East. bit.	West. subbit.	West. subbit.
Coal sulfur, % as fired	3.36	0.48	0.48
Coal ash, % as fired	15.1	6.3	6.3
Btu/lb, as fired	11,700	8,200	8,200
Sulfur emitted, % of total	92	85	85
Fly ash, % of total ash	80	80	80
NO_x emitted, ng/J	258	215	215
Boiler size[1], MW	500	500	500
Heat rate, J/kWh	10×10^6	11.1×10^6	11.1×10^6
Emission control			
NO_x control	SCR	SCR	SCR
NO_x reduction, %	80	80	80
SO_2 control	Limestone FGD	Lime spray dryer	Limestone FGD
Absorber trains[2]	5	4	5
Bypassed flue gas, %	0	12	28
SO_2 removal, overall %	89	65	65
SO_2 removal, absorber %	89	73	90
Fly ash control	Cold-side ESP	Baghouse	Hot-side ESP
Fly ash removal, %[3]	99.7	99.9	99.6

[1] Based on coal consumption and heat rate.

[2] Including one spare train.

[3] In collection device, excluding upstream fallout.

TABLE 7. CAPITAL AND ANNUAL COSTS OF SCR NO_x CONTROL ESTIMATED BY THE TVA/EPA STUDY /15/

	NO_x control	Total pollution control system $10^6	NO_x control, % of total
Capital ($/kW)			
Case 1	83.7	186.6	22
Case 2	100.2	166.7	30
Case 3	96.1	171.0	28
Annual (mills/kWh)			
Case 1	13.0	32.6	40
Case 2	15.8	28.9	55
Case 3	14.7	29.5	50

(Basis: 500 MW, 80% NO_x removal)

TABLE 8. ECONOMIC ANALYSIS OF FULL SCALE SCR SYSTEM APPLIED TO 500 MW COAL-FIRED UTILITY BOILERS (Analysis Based on EPRI Pilot Unit Test Results) /16/

RESULTS OF ECONOMIC ANALYSIS

Coal	Capital costs, $/kW	Annual costs, mills/kWh
Low sulfur	60 - 98	7.6 - 14.9
High sulfur	54 - 83	5.7 - 11.1

VARIABLES USED IN THE ECONOMIC ANALYSIS

Variables	Levels considered
Coal	Low sulfur western, high sulfur eastern
NO_x concentrations, ppm	200, 500, 800
NO_x removal, %	60, 80, 90

TABLE 9. COMPARISON OF TVA/EPA AND EPRI ECONOMIC ASSESSMENTS OF SELECTIVE CATALYTIC REDUCTION FOR NO_x CONTROL

Basis of Comparison

500 MW Utility Boiler
Low Sulfur Coal
80 Percent NO_x Control
1 Year Catalyst Life

	Capital costs, $/kW	Annual costs, mills/kWh
TVA/EPA study	100.2	15.8
EPRI study	79.1	11.7

TABLE 10. SCR APPLICATIONS IN THE STATE OF CALIFORNIA /17/

Location	Source (number of units)	Size MJ/sec	Fuel	NO_x removal %
Fletcher Refining	Boiler (2)	14.2 each	Oil and gas	50
Champlin Oil	Boiler (1)	--	--	--
Shell Oil Refinery	CO boiler (1)	--	--	--
USA Petrochem	Refinery heater (1)	19.0	Oil and gas	90
Tosco Refinery	Refinery heater (3)	35.2 total	Gas	90
Chevron Refinery[1]	Refinery heater (10)	--	--	--

[1]Proposed, not yet installed.

TABLE 11. COMMERCIAL APPLICATIONS OF FLUIDIZED BED COMBUSTION /10,11/

Utility	Station	New/ retrofit	Size, MW	Start-up
Northern States Power	Black Dog	Retrofit	125	1986
Colorado Ute	Nucla	Retrofit	100	1987
Tennessee Valley	Shawnee	New	160	1989

TABLE 12. STATUS OF PILOT UNIT DEMONSTRATIONS OF ADVANCED SO_x/NO_x PROCESSES /18/

Process	Status
Fluidized copper oxide bed	0.5 MW work is complete, larger scale tests are being considered
Spray dryer SO_x/electron beam NO_x	Testing will be completed by February 1985
SULF-X SO_x/NO_x	1.5 MW pilot unit is ready to begin testing
Electron beam SO_x/NO_x	6 MW pilot unit should start up in 1985